

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Juraj BABJAK et al.

Serial No. 10/729,911

Group Art Unit: 1742

Filed: December 9, 2003

Examiner: Melvyn J. Andrews

For: METHOD FOR REMOVAL OF Mn FROM COBALT SULFATE

SOLUTION

## DECLARATION UNDER 37. C.F.R. 1.132

HONORABLE COMMISSIONER OF PATENTS AND TRADEMARKS WASHINGTON, D.C. 20231

Sir:

Now comes Naoichi YAMAMOTO, a citizen of Suita-shi, Osaka-fu, Japan, who declares and states:

That I graduated from the Department of Science, Faculty of Chemistry in Kyoto University in the year 1963 and completed a postgraduate course of the Graduate School of Kyoto University in the year 1968.

That, after obtaining a Ph.D. degree in Science from Kyoto University, I worked in Kyoto University of Education as an assistant professor for 4.5 years, then in Kyoto University as an associate professor for 14.5 years and as a professor for 13.5 years in the field of research of solid

state chemistry.

That, under my direction and surveillance, the following experiments were implemented in the research laboratory of Seido Chemical Industry Company, Ltd. on October 5th and 12th, 2005.

# (1) Purpose of the experiments

The experiments was performed to investigate whether an addition of NaOH to NaOCl as an oxidative agent used for removing Mn from cobalt sulfate solution could result in the oxidation reduction potential (ORP) required for achieving a high degree of Mn removal.

# (2) Procedure of the experiments

Cobalt sulfate solution containing 100 g/L of cobalt and 25 mg/L of Mn having pH of 4 to 5 was prepared by dissolving the corresponding quantities of the respective carbonates in sulfuric acid.

To 2 L of the solution being heated to 50 °C was added 100 mL of an aqueous NaOCl solution having a concentration of 10 g/L in 30 minutes (Experiment 1).

In the comparative experiments, 100 mL of an aqueous solution of NaOCl (10 g/L) mixed with NaOH having a molar ratio of 1:1 and 1:2 was respectively added in 30 minutes into 2 L of the cobalt sulfate solution containing Mn at 50  $^{\circ}$  (Experiments 2 and 3).

The values of the ORP (mV) with respect to a standard hydrogen electrode (SHE) and pH of the solution were measured before and after

the oxidation process.

The resulting suspension was filtered and the concentration of Mn in the filtrate after removal of the precipitated Mn was determined.

## (3) Results

The results of the experiments were summarized in Table 1.

Table 1

Experiment No.		1	2	3
NaOCl : NaOH (molar ratio)		NaOCl only	1:1	1:2
pН	Start	4.8	4.0	4.6
	End	1.9	4.1	5.6
ORP (mV)	Start	690	855	790
	End	1260	850	660
Mn content in filtrate (mg/L)	Start	25	25	25
	End	0.1	19	25

According to the present application, a substantially complete removal of Mn contained in cobalt sulfate solution can be achieved by adding NaOCl as an oxidant to the solution of the pH of 2.5 to 6 to attain the ORP in the range of 1100 to 1300 mV (SHE), thereby oxidizing Mn to form a precipitate of Mn, and by removing the precipitate from the solution.

It is essential for this inventive method to use NaOCl to attain the ORP of 1100 to 1300 mV (SHE).

It was confirmed from the Experiment 1 that the addition of NaOCl alone to the solution lowered the pH to the preferable range (1.5 to 2.5), increased the ORP to the required level (1100 to 1300 mV), and thus achieved the desired degree of Mn removal.

In contrast, the Experiment 2 exhibited that the addition of the mixture of NaOCl and NaOH (molar ratio of 1:1) to the solution raised the pH, decreased the ORP, and thus caused an incomplete precipitation of Mn.

Further, the Experiment 3 showed that the addition of the mixture of NaOCl and NaOH (molar ratio of 1:2) not only decreased the ORP but also entirely failed to oxidize Mn.

The mixture of NaOCl and NaOH with a molar ratio of 1:2 is a reactant equivalent for the oxidation of Mn as follows;

 $MnSO_4 + NaOCl + 2NaOH \rightarrow MnO_2 + Na_2SO_4 + NaCl + H_2O$ 

However, the result of the Experiment 3 revealed that the addition of NaOH to NaOCl as the oxidant makes it difficult to oxidize Mn contained in the cobalt sulfate solution without oxidizing Co.

Thus, it was proved that the addition of NaOH to NaOCl does neither attain the ORP of the required range nor work as the oxidative agent for completely oxidizing Mn contained in the cobalt sulfate solution.

The undersigned Petitioner declares further that all statement made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Further declarant saith not.

Respectfully submitted,

Date: November 9, 2005

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Naoichi YAMAMOTO